

# Advanced LWR Nuclear Fuel Cladding System Development Trade-off Study

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Trade-off Study**

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**September 2012**

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## EXECUTIVE SUMMARY

The LWR Sustainability (LWRS) Program activities must support the timeline dictated by utility life extension decisions to demonstrate a lead test rod in a commercial reactor within 10 years. In order to maintain the demanding development schedule that must accompany this aggressive timeline, the LWRS Program focuses on advanced fuel cladding systems that retain standard  $\text{UO}_2$  fuel pellets for deployment in currently operating LWR power plants. The LWRS work scope focuses on fuel system components outside of the fuel pellet, allowing for alteration of the existing zirconium-based clad system through coatings, addition of ceramic sleeves, or complete replacement (e.g. fully ceramic cladding).

Through extensive literature reviews and conversations with stakeholders, the LWRS Trade-off Study team has identified preliminary fuel cladding technology performance criteria and categorized these criteria into three groups: critical performance criteria, desired performance criteria, and licensing criteria. Weights will be assigned based on input from stakeholders, nuclear industry experts, and the nuclear material science community. Advanced cladding technology options will be evaluated against one another based on these weighted parameters. Technologies will be selected for further development and testing based on the results of the evaluation. A discussion on the nuclear design trade-offs, accident performance, storage, economics, safety, and licensing capabilities offered by each competing technology is included in this Trade-off Study report as a basis for the performance criteria and technology selections.

The leading candidate technologies identified in this Trade-off Study include coated zircalloys, SiC/SiC, SiC CMC Zr-based alloy hybrids, advanced zircalloys, and engineered stainless steel alloys. Potential benefits and drawbacks have been identified in this study to aid in technology selection for further development and testing. The information provided in this Trade-off Study can be used in conjunction with continued input from industry stakeholders, national laboratories, and the U.S. Department of Energy-Nuclear Energy (DOE-NE) to identify technologies warranting further development and testing through the LWRS program.

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## ACRONYMS

ATWS	Anticipated Transient Without Scram
BDBA	Beyond Design Basis Accident
BWR	Boiling Water Reactor
CFR	Code of Federal Regulations
CFRCMC	Ceramic Fiber-Reinforced Ceramic Matrix Composite
CMC	Ceramic Matrix Composite
CVD	Chemical Vapor Deposition
CVI	Chemical Vapor Infiltration
DBA	Design Basis Accident
DOE	U.S. Department of Energy
DOE NE	DOE Office of Nuclear Energy
ECCS	Emergency Core Coolant System
EPRI	Electric Power Research Institute
F-M	Ferritic-Martinistic
GWd/tHM	Gigawatt day per ton heavy metal
GEH	General Electric Hitachi
IAC	Industry Advisory Committee
IASCC	Irradiation Assisted Corrosion Cracking
INL	Idaho National Laboratory
LEU	Low Enriched Uranium
LHGR	Linear Heat Generation Rate
LOCA	Loss of Coolant Accident
LPS	Liquid Phase Sintering
LWR	Light-Water Reactor
LWRS	Light Water Reactor Sustainability
LTR	Lead Test Rod
MI	Melt Infiltration
MWd	Mega Watt Day
MWt	Mega Watt Thermal
NITE	Nano-Infiltration and Transient Eutectic-phase
NPP	Nuclear Power Plant

NRC	U.S. Nuclear Regulatory Commission
PAN	Polyacrylonitrile
PCCI	Pellet Clad Chemical Interaction
PCI	Pellet Clad Interaction
PCMI	Pellet Clad Mechanical Interaction
PIP	Polymer Impregnation and Pyrolysis
PWR	Pressurized Water Reactor
PyC	Pyrolytic Carbon
QA	Quality Assurance
R&D	Research and Development
RIA	Reactivity Initiated Accident
SAFDL	Specified Acceptable Fuel Design Limits
SAMG	Severe Accident Management Guidelines
SBO	Station Blackout
SEM	Scanning Electron Microscope
SiC	Silicon Carbide
SWU	Separative Work Unit
T&FR	Technical & Functional Requirements
TMOL	Thermal Mechanical Operating Limit
UO <sub>2</sub>	Uranium Dioxide
U.S.	United States
UTS	Ultimate Tensile Strength
WEC	Westinghouse Electric Company
Zr-4	Zircaloy-4



# Advanced LWR Nuclear Fuel Cladding System Development Trade-Off Study

## 1. INTRODUCTION

The Advanced Light Water Reactor (LWR) Nuclear Fuel Development Research and Development (R&D) Pathway encompasses strategic research focused on improving reactor core economics and safety margins through the development of an advanced fuel cladding system. To achieve significant operating improvements while remaining within safety boundaries, significant steps beyond incremental improvements in the current generation of nuclear fuel are required. Fundamental improvements are required in the areas of nuclear fuel composition, cladding integrity, and the fuel/cladding interaction to allow power uprates and increased fuel burn-up allowance while potentially improving safety margin through the adoption of an “accident tolerant” fuel system that would offer improved coping time under accident scenarios. With a development time of about 20 – 25 years, advanced fuel designs must be started today and proven in current reactors if future reactor designs are to be able to use them with confidence.

### 1.1 LWRs Fuel Mission: Advanced Cladding

The LWR Sustainability (LWRS) Program activities must support the timeline dictated by utility life extension decisions (i.e. demonstration of a lead test rod in a commercial reactor within 10 years). In order to maintain the demanding development schedule that must accompany this aggressive timeline, the LWRS Program focuses on advanced fuel cladding systems that retain standard UO<sub>2</sub> fuel pellets for deployment in currently operating LWR power plants. The LWRS work scope focuses on fuel system components outside of the fuel pellet, allowing for alteration of the existing zirconium-based clad system through coatings, addition of ceramic sleeves, or complete replacement (e.g. fully ceramic cladding).

Through extensive literature reviews and conversations with stakeholders, the LWRS Trade-off Study team has identified preliminary fuel cladding technology performance criteria and categorized these criteria into three groups: critical performance criteria, desired performance criteria, and licensing criteria. Weights will be assigned based on input from stakeholders, nuclear industry experts, and the nuclear material science community. Advanced cladding technology options will be evaluated against one another based on these weighted parameters. Technologies will be selected for further development and testing based on the results of the evaluation. A discussion on the nuclear design trade-offs, accident performance, storage, economics, safety, and licensing capabilities offered by each competing technology is included in this Trade-off Study report as a basis for the performance criteria and technology selections.

### 1.2 Performance Criteria

The performance criteria identified in Tables 1-3 were constructed from review of nuclear industry reports, papers, and presentations with the intent of evaluating potential cladding technologies for further development and testing within the LWRS Advanced Nuclear Fuels Pathway. Each criterion will be weighted based on nuclear industry and stakeholder input and the criteria will be used to select technologies for further development and testing to support the LWRS Advanced Nuclear Fuels Pathway mission. A discussion on industry requirements and concerns used to construct the performance criteria is included in Section 2.0.

**Table 1. Identified critical cladding performance parameters which must be met for consideration in further development and testing.**

<b>CRITICAL Cladding Performance Parameters</b>	<b>Zr alloy (Standard)</b>	<b>New Technology Criteria</b>	<b>Comments</b>	<b>Ref #</b>
<b>Maximum Operating Temp (°C)</b>	650	>650	Temp where structure or oxidation/corrosion becomes limiting	1
<b>Neutron absorption cross section</b>	0.0142 cm <sup>-1</sup> 0.26 barns	<0.0142 cm <sup>-1</sup> <0.26 barns	Reduced parasitic absorption	1, 2
<b>U-235 penalty (% increase in U-235 required for equivalent 18 month cycle at constant nominal power)</b>	0	≤0	Equivalent fuel geometry	1
<b>Clad manufacturability (low cost; low complexity)</b>	Mfgr 3.05-4.57 x 10 <sup>6</sup> m/yr (1); \$20-\$30 per meter	Mfgr 3.05-4.57 x 10 <sup>6</sup> m/yr; (suggested acceptable increased fabrication costs of up to 50% given power uprates of 10-20%) (3)	Must be able to meet current mfg demand – highly complex technologies may be costly and time consuming to manufacture	4
<b>Coolant cladding chemical interaction/degradation</b>	Localized oxidation not to exceed 17% of cladding thickness before oxidation (5)	Not allowed under normal operating conditions or postulated accident conditions		1
<b>Creep (thermal or irradiation)</b>	Creep strain for Zircaloy-4 @ Room T: - 0.66% (7)	≤ Zircaloy	Can affect thermal conductivity, fuel clad interactions, and cladding structural integrity	1
<b>Operational lifetime</b>	62 MWd/kg U	> 80 MWd/kg U	U-total not U-235. Must be greater than Zr alloy standard	1, 3
<b>Lifecycle net cost of fuel system</b>	8.79 mills/kWeh	<8.79 mills/kWeh	(mill is a unit of measure = 0.001\$)	3
<b>High strength / ductility</b>	UTS 437 MPa for Zr4 @ Room Temp (7)	≥Zr std	UTS = ultimate tensile strength – averaged value between longitudinal and transverse strength	6
<b>Hermeticity</b>			Necessary to contain fission gases under nominal operating conditions	6
<b>Longer coping times during LOCA</b>	10 hours – dependent on the specific NPP (10 CFR 50.63 & RG 1.155)	>10 hours		8
<b>Hydrogen generation in LOCA (exothermic reaction rate in steam)</b>	Total H production not to exceed 1% of the total amount of H produced if all metal cladding were to react (5)	Minimal H reaction rate		
<b>Power uprates</b>	3411 MWt – typical LWR	10-20% increase		3

**Table 2. Cladding parameters which are desirable but not mandatory for consideration in further development and testing.**

Desirable Cladding Performance Parameters	Zr alloy (Std)	New Technology Criteria	Comments	Ref #
Melting Point (°C)	1843	>2000 desired	Must withstand accident scenarios	1
Density (g/cm <sup>3</sup> )	6.56	< 6.56	Leads to improved seismic response	1
Heat capacity	0.07 (cal/g/°C) 293 (J/kg/°C)	>0.07 >293	Increases ability of cladding to damp out temperature increases during accidents. Minor effect.	1
Unirradiated thermal conductivity (k) @300 °C (W/m K)	17.41	≥17.41		1
Clad swelling	Swelling occurs	≤Zircaloy	At reactor operating fluence and temperature	1
Fission gas release	1 failure/million fuel pins	≤ 1 failure/million fuel pins	At reactor operating fluence and temperature	4

**Table 3. Cladding parameters needed to obtain Nuclear Regulatory Commission (NRC) licensing.**

Licensing Criteria	Zr alloy (Standard)	New Technology Criteria	Comments	Ref #
Clad Peak Temperature Limit (°C) (NRC accident limit)	1200	Peak T Limit >1200		1
Departure from Nucleate Boiling Issues (PWR)	Not allowed	Not allowed	Increase in power density must be balanced with heat transfer area per volume	1
Storage/Transportation/Waste disposal			No new waste disposal challenges from activation products	9
Compatible with higher U-235 enrichment	5 wt % U	> 5 wt% U		9
Withstand longer cycle lengths	18-24 months	≥18 months	Less lost time due to outages	1
Chemical compatibility with LWR components				9
Consistently meet nuclear QA standards				6
Potential for demonstration as a non-fuel component and lead test rod in 5-10 year				4, 8

## 2. INDUSTRY REQUIREMENTS

Industry requirements for development of advanced fuel cladding concepts were compiled from various nuclear industry reports. A common emphasis from these reports was on accident tolerance while at a minimum maintaining current (if not better) levels of performance with limited negative impact on economics. In evaluating advanced material technologies, emphasis should be placed on both potential economic improvement as well as accident resistance in light of the Great Tohoku Earthquake and Tsunami that caused the crisis at the Fukushima Daiichi Nuclear Power Station in Japan. Many novel technologies show promise but may present concerns regarding industrial viability. A summary of the requirements identified are included in this section and support the compilation of performance criteria in Table. 1.

## 2.1 Economics

Any decision to pursue a new fuel design must be based on an analysis of the costs and benefits associated with the change. If the costs outweigh the benefits, there is little reason to incur the expenses associated with the change [10]. Economic considerations for advanced fuel technologies should include the cost/benefit ratios of increased nodal power (i.e. power uprates), higher fuel burnups, longer cycle lengths, manufacturability, reliability, and storage.

### 2.1.1 Increased Nodal Power (Power Uprates)

New fuel materials hold promise of supporting higher nodal powers associated with higher Thermal Mechanical Operating Limits (TMOLs) or Linear Heat Generation Rates (LHGRs) given improved thermal properties (e.g., thermal conductivity, or melting temperature). The fundamental fuel related Specified Acceptable Fuel Design Limits (SAFDL) associated with fuel melting requires that no melting occur during steady-state operation. For modern fuel, there is abundant margin to melt during steady-state operation and the maximum steady-state LHGR is dictated by the transient overpower which may be 30%, or more, depending on the nature of the transient. In addition to the SAFDL governing fuel melt, the maximum LHGR limit, must also conform to a cladding strain related SAFDL that is also dictated by the transient overpower. It is also important to consider the relationship between fuel thermal properties, fuel design and system response. While improved thermal conductivity may support higher LHGR limits, it may also result in a more severe transient response and may increase the likelihood of a coupled nuclear-thermal hydraulic oscillation (i.e., instability). The advanced fuel system concepts that take advantage of improved thermal properties to achieve a higher TMOL and nodal power should be considered in the context of all of the constraints imposed on the fuel and cladding. [11]

### 2.1.2 Burnup, Cycle Length, and Fuel Cycle Cost

Improved cladding technology should be designed to support increased heavy metal loading. Higher concentrations of U-235 per volume of fuel are needed to improve fuel cycle cost by improving U-235 utilization to produce fewer used fuel assemblies per unit of energy produced [1]. Increased smear densities will target  $>10 \text{ gm U/cm}^3$  to meet this requirement. As the uranium inventory increases, enrichment requirements are reduced and fabrication is reduced as fewer assemblies are required to achieve a constant burnup. All else being equal, a 10% increase in heavy metal loading would result in ~3% reduction in front-end fuel cost. As the U.S. Boiling Water Reactor (BWR) fleet favors Power Uprate and 24 month refueling intervals, decreasing heavy metal loading will likely result in some evaluated penalty (e.g. increased enrichment and cost, reduced cycle length or burnup). Conceptually, maximum enrichments could be increased beyond 5%; however, this would affect all aspects of the fuel cycle and so significant benefit would be needed to justify [11]. The main impact on  $>5 \text{ wt\%}$  enrichments is seen during the initial stages of the fuel cycle—particularly enrichment,  $\text{UF}_6$  shipping, conversion, and pellet fabrication. Significantly higher enrichments (up to 20 wt%) will require facility re-design, a new shipping container, and fabrication line re-design. After these stages, the fuel behaves in a manner similar to the current 5 wt% fuel, with the main impact being an increase in source terms and decay heat with higher discharge burnup. This results in a minimal impact on facility design for the later stages; the main impact will be a need to re-license the present facilities to take advantage of the benefits of the advanced fuel concepts. Raising the uranium enrichment will affect every part of the fuel cycle, from enrichment through storage of used fuel and, therefore, the impact on the fuel cycle needs to be considered [10].

A 24-month or longer fuel cycle is being proposed by the industry to reduce plant operating costs [1, 10, 3]. Current Zr-based alloy cladding is licensed to only 62 MWd/kgU. The limit on fuel rod burnup effectively limits the batch average discharge burnup to a range between 50 and 55 MWd/kgU. A longer lifetime of  $>90 \text{ MWd/kgU}$  equivalent is needed to meet the WEC requirements for advanced fuel

concepts [1]. Advanced cladding technologies will need to perform better than Zr-based alloys during longer fuel cycles and higher fuel burnup.

### **2.1.3 Manufacturability**

It is important that any alternative fuel system be capable of large scale industrial production (~3,000 to 10,000 tons per year) while meeting nuclear fuel quality standards on a consistent basis as large quantities of fuel rods are manufactured every year with essentially zero defects that could lead to primary cladding failure. The WEC report points out that in order to meet current industry demands, 10 to 15 million feet/year (3 to 4 1/2 million m/yr) of cladding must be manufactured. Increased complexity in manufacturing a given cladding design will tend to increase the cost of the cladding and increase the cost of fuel operation [1]. Some increases in fabrication cost can be absorbed without significant impact on total power generation cost as long as there is overall benefit (e.g. the overall economics are improved because of other fuel attributes, or significant safety benefit) [11]. With additional demand for advanced cladding materials, costs could be expected to be reduced in the future.

Manufacturing defects which range from primary hydriding of the cladding to weld contamination need to be considered. The new cladding may be expected to experience increased manufacturing related failures during early deployment. There are also significant cost and manufacturing barriers and the ability to hermetically seal ceramic cladding tubes after the fuel is loaded has yet to be demonstrated. Sourcing and manufacturing of an advanced Zr-based alloy would likely have similar cost to current Zr-based alloys, with the exception that it might be produced in lower volume (at least initially), preventing benefits that come with larger orders. It is important that any alternative fuel and cladding be capable of large scale industrial production, as large quantities of fuel rods are manufactured every year with essentially zero defects leading to primary cladding failure. Some increases in fabrication cost can be absorbed without significant impact on total power generation cost as long as there is overall benefit (e.g. the overall economics are improved because of other fuel attributes, or significant safety benefit).[11]

### **2.1.4 Reliability**

The following failure modes have been identified as having economic impacts on design of advanced fuel concepts [11] and should be considered when selecting advanced cladding technologies:

- Debris fretting related to the hardness of the cladding.
- Corrosion which is usually related to an inadvertent intrusion of aggressive chemicals into the reactor coolant. This is affected by the corrosion properties of the cladding.
- Pellet Clad Interaction (PCI) which is related to the cladding material's susceptibility to stress corrosion cracking & mechanical properties, the stress imposed upon the cladding by the pellet, the design of the fuel rod (e.g. inner clad) and the pellet (e.g. additives).
- Manufacturing defects which range from primary hydriding of the cladding to weld contamination. The new fuel forms may be expected to experience increased manufacturing related failures during early deployment.
- Post-failure degradation and fuel washout has a significant impact on the dose consequences of a primary fuel rod failure. Corrosion and hydrogen embrittlement are important considerations for the cladding material.

Advanced cladding system designs will assume failure criteria similar to that for standard clad fuel; specifically, failure implies loss of fission product containment from the pin. The current industry standard is approximately one failure per million fuel pins. This standard assumes a fuel burn-up normal to standard zircaloy/VO<sub>2</sub> on the order of 50-60 MWd/kgU. Some of the proposed advanced cladding designs could require higher enrichment nuclear fuel while also reducing or eliminating the hydrogen embrittlement and other neutron-irradiation-induced degradation issues associated with zircaloy clad. Hence, it is conceivable that substantially higher burn-ups and power uprates may be possible with

advanced cladding options. Given the potential performance enhancements associated with advanced cladding it may be reasonable to assume that a higher failure rate (per pin) would be acceptable. However, any increase in the allowed failure frequency would likely be less than an order of magnitude relative to the current standard and should be the subject of future systems analyses.

### **2.1.5 Used fuel storage and fuel cycle closure**

After discharge, fuel will reside in the fuel pool for some years before transferred to dry cask storage. The cladding material should not corrode under these conditions. It is noted that temperatures can be elevated during dry cask storage and the cladding material must be able to withstand this and also have acceptable creep characteristics & ductility so as to not experience undesirable deformation or cladding failure. Improvements in cladding performance may support higher storage temperatures, earlier storage after discharge and reduced requirements on the storage cask which could reduce storage costs. The stored energy in the fuel material will impact the temperatures experienced by the fuel in dry storage.

In addition to storage of discharged fuel, some consideration needs be given to the ultimate disposition of the fuel, including the potential for the closing of the fuel cycle in one manner or other. The President's Blue Ribbon Commission stated in the preliminary report that "There appears to be a widespread consensus that existing and reasonably foreseeable nuclear fuel cycle technologies would not eliminate the need for a geologic disposal facility or facilities; there also appears to be consensus that current and advanced fuel cycles should be analyzed as an interconnected system in which each element must be compatible with and support of the other elements." Fuel forms that have characteristics that aid in the recovery of fissile content would have some inherent advantages for such closure of the fuel cycle [11].

## **2.2 Accident Performance**

While accident resistance and risk reduction are highly desirable, they are not as easily quantifiable as other physical criteria. These are primarily addressed by changing the cladding to something that has a significantly reduced exothermic reaction with steam relative to zirconium-based alloys, enhanced mechanical strength at elevated temperatures and high thermal shock resistance. The concept of risk reduction in advanced cladding materials is even more technically challenging to quantify because it cross-cuts all the measureable physical properties. An AREVA Federal Services, LLC report [10] identifies risk reduction as "changes that lead to fewer fuel failures and fewer fuel mechanical problems".

General Electric Hitachi (GEH) identified the following safety concerns in an assessment report of advanced material options [12] for BWR reactors, but the same requirements can be applied to all current LWR reactors. For Design Basis Accidents (DBA), the primary considerations are related to the cladding as opposed to the fuel form (significant changes in melting temperature notwithstanding):

### ***Design Basis Accidents (DBA)***

1. Loss of Coolant Accident (LOCA) which primarily affects cladding integrity and is influenced by cladding temperature/strength/ductility at temperature, corrosion performance at high temperature, and ability to withstand the thermal loads imposed by Emergency Core Coolant System (ECCS) actuation. Fuel limits are set to protect both the cladding peak temperature (set by the characteristics of the cladding ductility) and the cladding reaction rate (considering both the operational corrosion as well as oxidation during the transient).
2. Reactivity Insertion Accident (RIA), or Control Rod Drop Accident for the BWR, is influenced by cladding ductility, as well as the thermal characteristics of the fuel form, such as fuel expulsion characteristics.
3. Fuel Handling Accident in which a fuel assembly is dropped onto other fuel assemblies in the spent fuel pool, which is affected by the ultimate strength of the cladding and fuel assembly mass.

### ***Beyond Design Basis Accident (BDBA)***

Fuel attributes that affect the consequences of BDBA events are:

1. High temperature capability of cladding and fuel.
2. Cladding resistance to chemical reactions under accident conditions.
3. The fuel should have acceptable interaction with water assuming the clad fails.
4. Hydrogen production or production of some other combustible or otherwise detrimental substance that can challenge pressure boundaries and containment systems.

In response to the Fukushima accident and the insights it brings to light, a Nuclear Regulatory Commission (NRC) Near-Term Task Force was formed to review the insights from the Fukushima Daiichi accident. Based on the review, the task force is recommending general and specific actions that it believes would be a reasonable, well-formulated set of actions to increase the level of safety associated with adequate protection of the public health and safety. The Task Force review pointed out that the NRC has come to rely on design-basis requirements and a patchwork of beyond-design-basis requirements and voluntary initiatives for maintaining safety. Design-basis requirements include consideration of anticipated operational occurrences and postulated accidents such as loss-of-coolant accidents. Voluntary initiatives have addressed some severe accident considerations, shutdown risk issues, containment vents for BWR Mark I designs, and severe accident management guidelines (SAMGs). The concept of beyond-design-basis requirements applies, for example, to as anticipated transient without scram (ATWS) and station blackout (SBO). The Commission's SBO requirements provide assurance that each nuclear power plant can maintain adequate core cooling and maintain containment integrity for its approved coping period (typically 4 to 8 hours) following an SBO. The NRC Task Force report concludes that revising 10 CFR 50.63 to expand the coping capability to include cooling the spent fuel, prevent a loss-of-coolant accident, and prevent containment failure would be a significant benefit. . [13]

## 2.3 Material Properties

AREVA Federal Services, LLC identifies the following high level physical criteria for evaluating advanced fuel concepts [10]. The criteria are consistent with many of the reports examined in the process of conducting the present trade study. Criteria, which apply to both the nuclear fuel and cladding, include:

- Chemical compatibility of the fuel and cladding with other LWR core components
- Melting temperature
- Thermal conductivity
- Thermal neutron cross section
- Manufacturability
- Gas retention
- Radiation performance

In a report to the U.S. Department of Energy (DOE) and industry stakeholders [12], GE Hitachi (GEH) Nuclear Energy points out that large scale conductivity degradation during irradiation of advanced cladding systems may have large effects on fuel performance and must be factored into the design and licensing process. The GEH report suggests that research continue into addressing hermeticity, irradiation activated creep, thermal conductivity changes with irradiation, and pellet-cladding mechanical interaction (PCMI) of advanced cladding technologies. For the development of advanced nuclear fuel cladding, the production of high steady-state power requires that the cladding perform in conjunction with the fuel form so as to prevent duty-related failures due to PCMI. The BWR and Pressurized Water Reactor (PWR) have very different water chemistries and need to be factored into the property assessment during development. For accident conditions, there is a concern for potential brittle failure modes, particularly when coupled with thermal shock resulting from the quenching that would occur when the emergency core cooling systems are initiated in cases where there could be core uncover. The challenge

to the ductility of components due to seismic and LOCA loads require additional supporting data and application assessment. In addition, since the material is viewed as being particularly attractive due to the perceived lack of detrimental interaction with water at all conditions, it will need to be confirmed that the reaction with water and potential for hydrogen generation is indeed eliminated or substantially reduced.[11]

Coating technology on Zr-based alloys will need to demonstrate that the coatings will not spall off during operation, and that any cracking or crazing of the coating doesn't negatively impact the benefits of the coating or the performance of the underlying fuel rod cladding. Fuel rod insertion into the fuel bundle may scrape away the coating locally, and the effects of this type of scratch on performance should be evaluated. Also, the method of applying the coating must consider the optimized process utilized to fabricate current Zircaloy-based cladding to avoid any negative impacts. [14]

Post-failure degradation and fuel washout has a significant impact on the dose consequences of a primary fuel rod failure. Corrosion and hydrogen embrittlement are important considerations for the cladding material, and compatibility with water is a consideration for the fuel form [11]. Delayed hydride cracking is another concern with Zr-based alloys [16] and should be addressed in the development and testing of advanced Zr-based alloys. Improvement to corrosion resistance and to hydrogen-related properties (such as ductility) would improve fuel rod failure resistance under storage conditions.[11]

## 2.4 Licensing Requirements

The evaluation of fuel performance expectations needs to be measured against the NRC licensing basis requirements and potential for benefits to the entire core and reactor system. The fuel qualification process traditionally involves a combination of fuel design, fabrication process definition and fuel performance qualification, using in-reactor testing and performance analysis. The qualification process requires the development of several interrelated items, including:

- Choice of reactor type(s) that will use the product.
- Develop technical and functional requirements (T&FR) and a fuel system specifications; this may then require adding quality assurance (QA) steps to the fabrication process. The T&FR and specification can be rough and should be broad enough so as to require as few changes as possible in the future. Changes to these requirements may affect the subsequent processes in qualification and require back-stepping to correct.
- Description of the fabrication process that will produce a uniform product. Conceptual design of engineering-scale and full-scale fabrication processes to allow assessment of efficiency loss (for example, batch yield or uranium losses), capital cost, and production cost.
- Prepare a Performance/Safety case. The initial performance/safety case will serve as a gap analysis to uncover which fuel performance issues require further analysis/modeling and/or testing. The initial draft may also reveal where design changes are required.

The description of general fuel system qualification timeline indicates ~23 years from initial design to completion of the qualification process. The first nine years, however, are consumed with building and testing prototypes. The fuel system design down-selection does not occur until year nine. It may be possible to replace these initial nine years with a much shorter period of analysis and design refinement, with some benchmark experimentation, still resulting in a final lead test rod design. In-reactor testing should be designed to address issues that cannot be satisfactorily analyzed or modeled, or to validate the results of modeling. [4]



### 2.4.1 General Design Criteria

The IAEA Safety Guide [16] provides a comprehensive description of fuel system designs considerations, many of which have been identified in this report. The following is a summary of the design considerations from the IAEA guide pertaining to cladding materials for BWR and PWR reactors:

- Thermal hydraulic limits such as maximum linear heat generation rate, the minimum critical power ratio, the minimum departure from nucleate boiling ratio, the peak fuel temperature and the cladding temperature should be set in such a way that there are sufficient margins in operational states to keep the failure rates of fuel elements under design basis accident conditions to an acceptably low level.
- Changes in the thermal conductivity of the pellets and thermal conductance of the gap between pellet and cladding due to burnup dependent effects such as oxide densification, swelling, accumulation of fission products and other changes in the microstructure of pellets.
- Changes in the composition and microstructure of the fuel due to burnup
- Effects of gaseous fission products on the internal pressure of a fuel element and the thermal conductance of the pellet-to-cladding gap.
- The corrosive effects of fission products on the cladding.
- Leak-tight cladding for all operational states throughout its lifetime.
- Effects of temperature, pressure, irradiation, fission products, static and dynamic mechanical loads, including seismic loads, flow induced vibration and changes in the chemical characteristics of the constituent materials.
- Static and dynamic mechanical loads, including thermal stress, acting in operational states and in design basis accident conditions.
- Changes in mechanical properties (strength, creep and stress relaxation) and changes in the corrosion related behavior of the cladding with temperature.
- Cladding stress and strain limits for different operational states.
- Consequences of reactor depressurization events (in normal operation and following anticipated operational transients such as those initiated by the automatic depressurization system) in terms of the potential for failure of the cladding.
- Effects of fast neutrons on fuel assemblies (including control devices and burnable poisons), on metallurgical properties such as the tensile strength of the cladding, ductility and creep behavior, fuel densification and swelling (in radial and axial directions), and on the geometrical stability of all materials.
- Effects on the integrity of the fuel cladding of local and global power transients due to fuel shuffling, movements of control devices or other reactivity changes.
- Minimize stress corrosion cracking induced by pellet-cladding interactions in the presence of fission products.
- Mechanical loading of the cladding due to the length of unsupported plenum.
- Potential adverse effects of burnable poisons in the fuel on its thermal properties, and on the chemical, mechanical and metallurgical properties of the fuel and cladding material.
- Compatibility with the coolant environment in all operational states, including shutdown and refuelling.
- Oxidation or other chemical changes tending to lead to the formation of deposits on the surface of the cladding which may affect the transfer of heat from the fuel element.
- Limited hydrogen content of zirconium alloy cladding to reduce the likelihood of fuel defects being caused by the embrittlement of cladding during operation.
- Vibration and fretting caused by coolant flow.
- Postulated initiating events such as a loss of coolant accident.

- Limited bowing and deformation of fuel elements.
- Strain fatigue should not be able to cause the failure of a fuel assembly.
- Withstand the mechanical and hydraulic hold-down forces without unacceptable deformation.

## **2.4.2 Determining Enrichment Limits**

The fuel cycle should be designed with appropriate levels of enrichment and appropriate means of controlling the core reactivity and the power distribution in order to safely extract energy from the fuel in the most economic manner.. Excess reactivity is needed to operate the reactor until the end of a fuel cycle and/or to adjust the power level of the during a fuel cycle. Boron is used in LWRs to absorb the excess core reactivity. Its concentration can be reduced for overriding a xenon transient and re-establishing the core power levels. A specified margin to prevent criticality in the storage of dry fuel and in the storage pool should be incorporated in determining the maximum fuel enrichment. Changes in properties of the fuel pellets and the cladding in conditions of high fuel burnup may affect the behavior of fuel elements in accident conditions such as reactivity initiated accidents or loss of coolant accidents, therefore this should be considered in the cladding design. [16].

## **2.4.3 NRC Acceptance Criteria**

The United States NRC Part 50 (Domestic Licensing of Production and Utilization Facilities) defines the following criteria for light water reactors in terms of the emergency core cooling systems [5]. This criteria should be considered when designing advanced cladding systems:

- The calculated maximum fuel element cladding temperature shall not exceed 1200 °C (2200° F) during emergency cooling conditions (as applied to current Zr-based alloys).
- Calculated total oxidation of the cladding shall not exceed 0.17 times the total cladding thickness before oxidation.
- Total amount of hydrogen generated from the chemical reaction of the cladding with water or steam shall not exceed 1% of the hypothetical amount that would be generated if all of the metal in the cladding cylinders surrounding the fuel, excluding the cladding surrounding the plenum volume, were to react.

# **3. ADVANCED CLADDING TECHNOLOGIES**

Advanced cladding technologies examined are comprised of coatings on existing zirconium-based cladding, hydrid ceramic/metal cladding, or complete replacement of the conventional nuclear fuel clad material. The potential benefits of each of these technology categories is varied, from simply reducing the rate of fretting failure to allowing operation at higher temperatures with larger safety margins. Table 4 summarizes the potential benefits and drawbacks of specific technologies identified in this Trade-off Study.

**Table 4. Cladding technologies considered in the LWRS Trade-off Study along with potential benefits and drawbacks for each.**

Type	Technology	Potential Benefit	Potential Drawback
<b>Zr-alloy Coatings</b>	SiC	Enables adequate thermal characteristics and compatibility with UO <sub>2</sub> fuel for use in higher heat rate applications. Retains the small/no fuel clad gap characteristic yielding good overall thermal conductivity of zirconium cladding	Cannot withstand the same temperature range of full SiC cladding. Yield strength limited (650 C). Minimal development experience.
	Ti <sub>3</sub> AlC <sub>2</sub>	MAX-phase coating. High melting point (2000 C), high thermal conductivity (40 W/mK), high strength and fracture toughness, corrosion and oxidation resistant, elastically rigid	Minimal development experience. Oxidation Limited (1450 C)
	Ti <sub>3</sub> SiC <sub>2</sub>	MAX phase coating. High melting point (2000 C), high thermal conductivity (37 W/mK), high strength and fracture toughness, corrosion and oxidation resistant, elastically rigid	Minimal development experience. Oxidation Limited (1000 C)
	General coating technologies	Easy application of coatings onto Zr-based alloys; maintains same base material as current cladding (i.e., no hermeticity issues); avoids new PCMI/PCCI issues.	Coatings could be scraped off during fuel assembly; localized region of exposure could cause rapid oxidation of underlying Zr-based alloys.
<b>Hybrid</b>	SiC CMC Zr-4	High temperature strength, low chemical reactivity, no exothermic reaction, very hard material prevents fretting failure, retains the small/no fuel clad gap characteristic. Zr-4 inner tube provides hermetic seal to fuel.	Hydrogen generation from the inner Zr-4 tube under LOCA – potential hydrogen embrittlement. Bonding techniques between SiC CMC and Zr-4 tubing need to be developed and tested.
<b>Fully Replacement Tubing</b>	SiC/SiC	High temperature strength, low chemical reactivity, no exothermic reaction, very hard material prevents fretting failure. Enables adequate thermal characteristics and compatibility with UO <sub>2</sub> fuel for use in higher heat rate applications.	Requires a large fuel-cladding gap that does not integrate well with the poor thermal performance of UO <sub>2</sub> . Joining technology for end cap seals (to create hermetic seal) has not been well developed and tested.
	Ti <sub>3</sub> AlC <sub>2</sub>	Superb machinability with high degree of toughness. High melting point (2000 C), high thermal conductivity (40 W/mK), high strength and fracture toughness, corrosion and oxidation resistant, elastically rigid	Minimal development experience. Oxidation Limited (1450 C). High thermal neutron absorption cross section, comparable to SS.
	Ti <sub>3</sub> SiC <sub>2</sub>	Superb machinability with high degree of toughness, high melting point (2000 C), high thermal conductivity (37 W/mK), high strength and fracture toughness, corrosion and oxidation resistant, elastically rigid	Minimal development experience. Oxidation Limited (1000 C). High thermal neutron absorption cross section, comparable to SS.
	Engineered SS	Wide range of engineered alloys currently being researched. Increase in Cr and Ni content in austenitic steels enhances oxidation resistance. Alumina-forming ferritic alloys exhibit exceptional oxidation resistance. Superb machinability and weldability. High temperature capability. Water resistance in steam. High corrosion resistance. High temperature strength. High toughness compared to Zr-based alloys.	High thermal neutron absorption cross section
	Advanced Zr-based alloys	Most easily assimilated into the current licensing methods and regulatory framework due to similarity and improvement upon existing Zr-based alloys; anticipated reduction in hydrogen pickup and/or increased hydrogen solubility to reduce formation of zirconium hydrides.	The likelihood of further improvements making significant difference is limited.

### **3.1 Zirconium-based Cladding (Standard)**

In the 1950s development of nuclear propelled naval submarines prompted the selection and development of a cladding material having low neutron absorption cross-section, high strength and good corrosion performance in hot water. Six decades of active alloy development has produced tailored alloy chemistries and processing methodologies that provide an adequate measure of corrosion behavior under pressurized or boiling water reactor (PWR or BWR) conditions while limiting irradiation growth and creep to the extent that these phenomena are now frequently inconsequential to reactor operation. These attractive properties of zirconium alloys render them well suited for use as nuclear fuel cladding and structural components in conventional LWR oxide fuel bundles. The satisfactory performance of zirconium alloys is challenged once a shift is made from an environment associated with normal operating conditions in LWRs to reactor accident scenarios. A variety of accident sequences can result in the loss of cooling capability inside the core and loss of coolant that will eventually drive up the fuel temperature and expose the cladding to a high-temperature steam environment [4].

#### **3.1.1 Corrosion**

Zirconium alloys in general are highly resistant to corrosion; however, they are not immune to oxidation in the aggressive conditions that exist inside a commercial nuclear reactor. The corrosion issues for zirconium alloys in Boiling Water Reactors (BWRs) and Power Water Reactors (PWRs) are unique due to the differences in operating conditions and alloys employed. BWRs utilize Zr-2, while PWRs previously used Zr-4 and are now transitioning to Zr-Nb cladding (Zirlo™ and M5™). Other major differences between the reactor types that affect corrosion are coolant boiling in BWRs, high concentration of hydrogen in PWR coolant, high concentration of oxygen in BWR coolant, and higher PWR operating temperature. Corrosion is currently a design-limiting issue in LWRs and is extremely complex, and today remains poorly understood. [4]

#### **3.1.2 Oxidation**

For oxidation kinetics of zirconium alloys in high-temperature steam environments the formed non-stoichiometric to stoichiometric zirconium oxide layer remains coherent, and parabolic oxidation kinetics are observed for long periods of time. Because the oxidizing species is water (steam), the reaction produces a significant amount of hydrogen gas. The rate of heat production due to the oxidation reaction in the cladding becomes significant at temperatures above 1200°C. At this point the oxidation reaction has the potential to exceed decay heat production in the fuel to become the dominant source of fuel temperature rise. This self-catalytic reaction quickly drives up the temperature in the fuel and results in oxidation of the entire cladding, converting the clad to the brittle ceramic ZrO<sub>2</sub>. Rapid oxide layer growth and increased solubility of oxygen in the β-Zr phase at temperatures above 1200°C that result in loss of ductility in the cladding are the basis for the regulatory criteria pertinent to a design basis LOCA (10CFR50.46) that limits the maximum cladding temperature to 1204°C (2200°F). Accordingly, the maximum extent oxidation in the cladding is limited to 17% of its initial thickness. [4]

#### **3.1.3 Hydrogen Pick-up**

Hydrogen pick-up is the absorption into zircaloy of hydrogen generated during the low rate surface corrosion process on the zirconium. The oxidation of zirconium by water generates free hydrogen ions which can then permeate into the zirconium metal. The solubility of hydrogen is extremely low at LWR operating temperatures (80-100 ppm); as a result, hydrogen precipitates out as hydrides. These hydrides are deleterious to the corrosion properties, have a higher coefficient of thermal expansion thus effecting local dimensional stability and stress and ultimately the mechanical properties of the zirconium alloys. The hydrides precipitate and then migrate to areas of high stress, which can result in delayed hydride cracking. The presence of hydrides also causes increased uniform corrosion rate, although the mechanism for this increase is not well understood. Additionally, due to the low density of the hydrides, hydrogen

pick-up causes local swelling in the zirconium alloys. Another concern with the presence of hydrides is their effect on long term stability of the cladding during long term dry storage. [4]

### **3.1.4 Dimensional Stability**

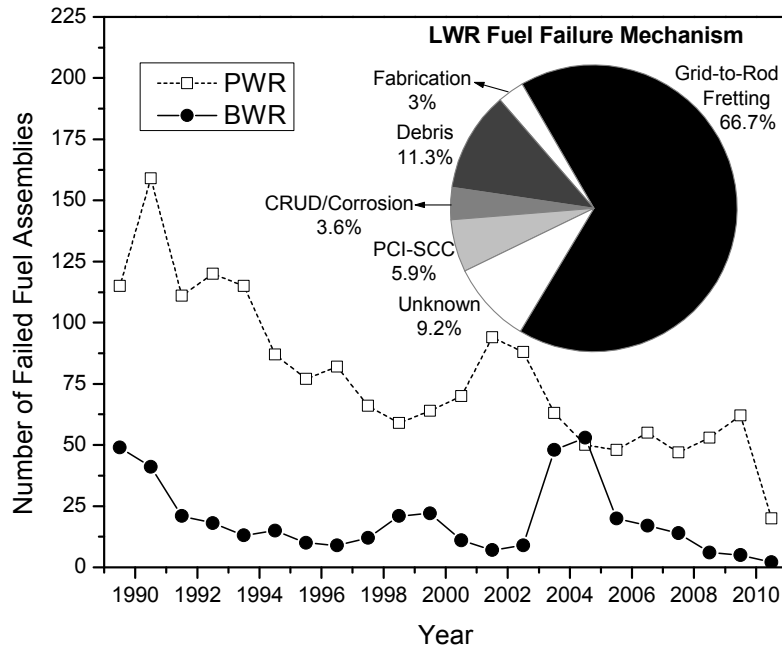
Dimensional stability is critical for reactor components, which are designed to tight tolerances. Deformation can lead to fuel channel and fuel assembly bowing which reduce thermal margin in plant design and can result in grid-to-rod vibration referred to as “grid fretting.” Dimensional instability in zirconium alloys is due to hydride volume changes, irradiation growth due to the hexagonal close-packed (HCP) structure and irradiation creep (thermal creep is insignificant at operating temperatures).

Irradiation creep is critical to the interaction of the cladding with the fuel pellets. Initially a gap exists between the fuel pellet and cladding; the cladding then creeps down to close this gap. At higher burn-ups (>50 GWd/tHM) the gap begins to reopen due to fission gas pressure. Creep is also the limiting mechanical property for many accident scenarios, such as a LOCA, due to the high temperatures seen during these accidents. Zirlo™ and M5™ have improved irradiation and thermal creep properties, allowing a larger safety margin during accident scenarios [4].

### **3.1.5 Zirconium Alloy Failure Rates**

The nuclear industry has made great strides in understanding the zircaloy/ $\text{UO}_2$  fuel system with systematic improvement in performance as measured by failed assemblies. This is evidenced by inspection of the timeline in Figure 1, which shows the impressive improvement in performance (reduced failure of assemblies) for both PWR and BWR systems. Figure 1 also includes the current modes of pin failure, indicating grid-to-rod fretting as the major contributor. Note that PCI-SCC refers to pellet-clad interaction stress corrosion cracking.

Advanced cladding system designs will assume failure criteria similar to that for standard clad fuel; specifically, failure implies loss of fission product containment from the pin. The current industry standard is approximately one failure per million fuel pins; this rate will be adopted as the maximum allowable failure in the development of advanced cladding under the LWRS Fuels Pathway. This standard assumes a fuel burn-up normal to standard Zr-alloy/ $\text{UO}_2$  on the order of 50-60 MWd/kgU. Some of the proposed advanced cladding designs could require higher enrichment nuclear fuel while also reducing or eliminating the hydrogen embrittlement and other neutron-irradiation-induced degradation issues associated with zircaloy clad (e.g. silicon carbide cladding designs). Hence, it is conceivable that substantially higher burn-ups and power uprates may be possible with advanced cladding options. Given the potential performance enhancements associated with advanced cladding it may be reasonable to assume that a higher failure rate (per pin) would be acceptable. However, any increase in the allowed failure frequency would likely be less than an order of magnitude relative to the current standard and should be the subject of future systems analyses [4]



**Figure 1. Timeline reflecting improved performance in commercial fuel assemblies and breakdown of current pin failure mechanisms in the industry [4].**

### 3.2 Silicon Carbide (SiC)

The use of silicon carbide (SiC) materials in cladding designs is promising for improved performance in certain areas under LWR core conditions, particularly under conditions where properties of zirconium based alloys could be significantly degraded, such as LOCA. SiC-based cladding for nuclear fuel may provide larger safety margins during transients and high burnup capability (>62 MWd/kgU) with longer cycle lengths or uprated operation (i.e. higher heat fluxes) [3]. SiC-based cladding may also be useful for advanced reactor concepts that require high fuel coolant temperatures such as superheated coolant or high-temperature gas reactors where physical properties of metals would be degraded. Unlike zirconium based alloys, SiC-based materials would retain its strength and will not creep up to 1300 °C and remains viable to even higher temperatures until onset of fuel-clad reaction. SiC has been shown to be stable to extremely high irradiation doses after the initial irradiation effects, which include swelling and changes to strength and thermal conductivity, are saturated after a few months of typical operation. There is also a neutronic benefit, as SiC materials parasitically captures fewer neutrons than Zr-based alloys, has very low activation and contributes a little more to neutron moderation [3].

Nuclear applications of SiC have been under investigation for over 50 years [12]. The majority of the research has focused primarily on the monolithic shell of SiC in TRISO fuel for gas-cooled reactors, but this research forms a basis for many of the properties important to LWR cladding. A relatively large amount of work has also resulted from experimental and analytic investigations of SiC composites as the first wall in potential fusion reactors [12]. These tests contributed to the development of radiation-resistant materials and provide part of the technical basis for SiC-based composites for potential LWR applications.

In an Industry Advisory Committee (IAC) to the Idaho National Laboratory Advanced Light Water Reactor Fuel Development Program status report (IAC) report [14], the IAC noted that based on current SiC-only clad concepts, SiC cladding would require increased wall thickness relative to Zr-based alloys. Given a constraint to maintain the outer fuel pin diameter, increased cladding volume results in a corresponding loss of 20% fuel volume such that SiC-clad fuel would require higher enrichments. This effect is somewhat offset by the neutron absorption cross-section for SiC, which is 25% that of Zr-based

alloys. The lack of a robust joining method for cladding is also a current development issue for SiC-only cladding concepts, as is the intrinsic low impact toughness of the base material.

### 3.2.1 SiC Ceramic Matrix Composites (CMCs)

SiC CMC is a very strong, high temperature ceramic material that is also chemically nonreactive. Very fine filaments (~ 10 micron) are combined into fiber tow which are then woven or braided into a tubular cladding form to enhance the SiC cladding structural strength while mitigating the brittle nature of monolithic SiC. The reinforcing fibers are bound into place by an additional bulk silicon carbide matrix designed to prevent the weave from moving under stress. A very thin interlayer of ductile high temperature binding material between the fibers and the matrix allows the ceramic materials to flex and twist without causing brittle failure.

The process of forming a thin-walled CMC tube uses textile methods of continuous fiber braid lay-up (preforming) or filament winding over a mandrel followed by formation of a very thin (sub micron) interface layer between the fibers and adjacent ceramic matrix followed by the process to form the SiC ceramic matrix. The interface layer between the fiber and the matrix is deposited for the purpose of transferring mechanical load within and through the ceramic fiber reinforced CMC (CFRCMC). This layer can consist of a number of materials such as pyrolytic carbon (PyC), oxide ceramics, or boron nitride (BN). PyC has known radiation stability issues that lead to cracking, such that it may not be appropriate for fabrication of SiC components intended for reactor applications.

There are multiple industrial processes for forming the SiC ceramic matrix surrounding the continuous ceramic fibers. The most common processes include:

- Chemical Vapor Infiltration (CVI) of the SiC (using isothermal or temperature-gradient and forced-flow, isobaric or pulsed flow methods),
- Pre-ceramic liquid polymer impregnation and pyrolysis (PIP) formation followed by elevated temperature conversion to SiC,
- Direct reaction-formed SiC matrix using melt-infiltration (MI) methods,
- Nano-Infiltration and Transient Eutectic-phase (NITE) formation of the SiC matrix using the transient liquid phase-assisted pressure sintering process.

In each process the resulting local chemical bond between Si to C is the same. However, each process needs to be controlled to the desired crystalline phase(beta or alpha) and to achieve a Si/C ratio equal to 1. Variations in the local Si/C ratio (greater than or less than 1, free Si or free C) can affect material properties.

**Chemical Vapor Infiltration (CVI)** is a variant on chemical vapor deposition (CVD). CVD implies deposition onto a surface, whereas CVI implies deposition within a body. The CVI process uses reactant gases that need to diffuse into an isothermal or temperature-graded porous fiber preform and form a deposition. The deposited SiC material is a result of local chemical reactions occurring at or on the fiber surfaces forming SiC. The infiltration of the gaseous precursor into the preform is driven by either diffusion processes or an imposed external gas pressure. The infiltration proceeds as the silicon carbide (matrix) deposition fills the space between the fibers, forming composite material in which the SiC matrix is the deposited material and the fibers of the preform make up the dispersed phase. To make the completed CMC composite, an inter-layer of carbon is also needed between the SiC fiber and the SiC matrix. This layer is made using a hydrocarbon precursor (such as CH<sub>4</sub>). The matrix densification stops when the preform surface pores are closed. The final residual closed porosity of the ceramic composites fabricated by the CVI method may reach 10-15% for a typical two-dimensional fabric lay-up architecture. By light machining of the surface additional vapor penetration into the fibers can be effected into the matrix.

CVI is a “batch” process known to require capital intensive and complex reactors, costly reactant gases, and control of potential flammable off-gases. Process run times can range from days to weeks, yielding a low final part throughput. Any stoppage of the CVI process can result in an internal Si or C rich layer over the deposited SiC layer. The presence of this layer needs to be assessed for quality and performance reasons, as either free Si or C would result in poor performance within a radiation environment. The CVI process relies on SiC formation from specific gas phase reactants which can be adjusted during SiC matrix formation to yield layers of SiC containing silicon or carbon rich layers. In general, for nuclear applications where the CFRCMC will be exposed to neutron radiation, a stoichiometric composition ( $\text{Si/C} = 1$ ) is preferred, as excess Si or C can lead to local swelling under neutron irradiation [4].

**Polymer Impregnation and Pyrolysis (PIP)** uses liquid polymers which will convert to form the desired ceramic matrix for CFRCMC. Current PIP techniques can be used to form ceramic matrices consisting of carbon, silicon carbide (SiC), silicon oxycarbide (SiOC), silicon nitride ( $\text{Si}_3\text{N}_4$ ) and silicon oxynitride ( $\text{Si}_3\text{ON}_3$ ). PIP is a low temperature liquid polymer method, which can potentially allow low cost production of simple and complex parts including CFRCMCs. PIP is performed at room temperature and with conventional industrial processing equipment, including use of high vacuum to aid in polymer impregnation into the fiber preform and radiant heating furnaces to cure and transform the polymer into nano SiC which can sinter at higher temperature into SiC grains. With the exception of the pre-ceramic polymers used, the PIP process is applicable to large L/D tubular products with scale-up to 10 feet and longer demonstrated. However, early work conducted in the mid 1990’s using a previous lower purity version of the polymer had been demonstrated to be unstable in a radiation environment. The primary reason for the radiation instability is the nano-crystalline siliconoxycarbide structure that progressively crystallizes during irradiation at temperatures above the amorphization-threshold temperature for SiC ( $\sim 150^\circ\text{C}$ ), accompanying substantial volumetric contraction and embrittlement due to extensive intergranular micro-cracking. [4].

Improving the irradiation stability of PIP SiC matrix composite is considered possible by increasing the purity of the starting polymer, increasing the polymer yield by adding additional cross linked chains containing Si, minimizing the amount of polymer-derived SiC in the matrix by loading the polymer precursor with filler particulates such as SiC, and/or transforming the PIP matrix into a fully crystallized and stoichiometric form of SiC through a heat treatment at temperatures exceeding the conventional pyrolysis temperature. Limited effort toward development of the fully crystallized PIP SiC matrix has revealed a technical processing challenge of preventing matrix damage upon crystallization. Therefore, research and development will be required for the development of PIP SiC/SiC composites that meet the baseline properties requirements for the LWR fuel application, to be followed by more extensive evaluation including the neutron irradiation and environmental effects upon successful development of the basic process [4].

**Melt Infiltration (MI)** fabrication of the SiC matrix would involve filling the pores in the composite preform by the liquid reaction between molten silicon and carbon to form silicon carbide. The MI process needs to be controlled as there is an exothermic temperature rise during the liquid silicon-carbon reactions and a local expansion (less than 2%) during formation of SiC bonds. In general, with silicon-based MI composites the upper use application temperature may be limited to the melting point of any free silicon remaining in the composite. SiC-fiber reinforced, melt-infiltrated SiC matrix composites are the leading candidate materials for aircraft and land-based turbine engine applications such as a combustor liner. However, the MI method results in significant residual silicon metal which may not be compatible for use long term use in a neutron environment. Free silicon is known to swell under neutron irradiation unless the composite is used at elevated temperature where any radiation induced defects would be annealed out [4].

The **Nano-Infiltration and Transient Eutectic-phase (NITE)** Process makes use of powder sintering for the matrix densification in SiC matrix composites. The sintering method adopted in the



NITE process is a liquid phase-assisted pressure sintering using nano-phase SiC powder mixed with small amounts of oxide additives. The NITE process is distinguished from the conventional liquid phase sintering (LPS) of SiC in that the resultant material is primarily the polycrystalline SiC (beta or alpha) with a small amount of oxide remaining in multi-grain junctions; whereas, the conventional LPS SiC is itself a composite material consisting of re-precipitated SiC grains embedded in the oxide matrix. Similar to the MI and PIP processes, the reinforcing fibers are coated with the protective debond interlayer of PyC prior to the matrix formation by the NITE process. The NITE process can use only Tyranno™-SA3 and Sylramic™ among the commercial near-stoichiometric SiC fibers because of the high processing temperature of ~1800°C. The NITE SiC/SiC composite has proven to be tolerant against neutron irradiation at temperatures and fluence levels to which it has been evaluated. No report has been published regarding the chemical stability of NITE SiC/SiC in the LWR coolant environment. Steam corrosion of the NITE matrix material is reportedly comparable with high purity CVD SiC, although differences in the oxidation mechanism are implied. Technology for producing thin-walled small diameter tubular components is not established for the NITE SiC/SiC [4].

### **3.2.2 SiC/SiC Cladding**

The IAC status report [14] describes an all SiC/SiC ceramic fuel rod concept. The basic structural component of the proposed cladding is a layer-pair consisting of a SiC CMC outer layer for strength and a dense monolithic  $\beta$ -SiC inner layer for impermeability. Monolithic SiC is stable to high temperatures and high levels of neutron flux, and exhibits good thermal conductivity, however monolithic SiC suffers from brittle fracture. SiC/SiC composites offer improved fracture toughness and retain the high temperature and irradiation stability of monolithic SiC. Due to the complex structure typically containing porosity and interfaces, the thermal conductivity is reduced, therefore it is important to examine variations in composite structure and processing that affect thermal conductivity [15]. The SiC/SiC concept described in the IAC report would utilize a cladding tube with sealed SiC end-caps where the external dimensions are the same as for current LWR rods. The tube would initially be fabricated with one sealed end-cap. The open end would then be closed by a hermitically sealed end cap after loading the fuel pellets.

The thicker SiC/SiC cladding wall, compared to Zr-based cladding, will require increased enrichment fuel. This increase in fuel enrichment in conjunction with the need for a robust joining method, are concerns with the all SiC/SiC designs, as previously discussed. In addition, SiC/SiC designs require a dense, impermeable layer while avoiding flaws and cracks resulting in tight control of the very complex manufacturing processes. It is expected that the SiC/SiC cladding design will be more resistant to fretting in normal operation; however, erosion and corrosion may be an issue. Non-stoichiometric SiC-based materials have demonstrated even more significant mass loss, whether by corrosion or erosion [4]. The presence of free silicon, or presumably other sintering aids residing at grain boundaries, has previously been shown to enhance corrosion of monolithic SiC for water temperatures as low as 290°C [4]. The mass loss, beyond any concerns regarding irradiation instability underlying the material loss, could raise issues as the very hard SiC particulates (or possibly SiO<sub>2</sub>) are transported through the coolant to heat exchangers and pumps. It is also conceivable that an irradiation-assisted-corrosion process that enhances the surface reaction may occur [4]. These processes in SiC/SiC should be compared to same processes for Zr-based alloys to determine potential performance impact.

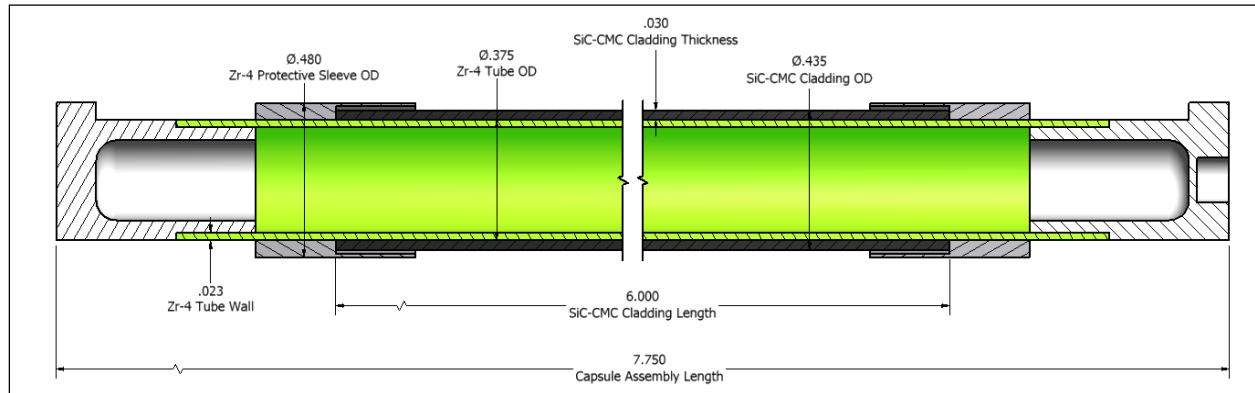
The SiC/SiC design is expected to significantly extend operation in severe accident conditions (increased system coping time) with significant reduction in hydrogen production, however it is not clear how the material will perform in impact scenarios, such as a dropped fuel assembly or seismic events. Additionally, it is not clear how the material will perform in normal operating conditions if subjected to assembly distortion or fuel rod bow behaviors. Additional research is also needed to evaluate potential fission product reactions and Pellet Clad Chemical Interaction (PCCI) [14].

### 3.2.3 SiC Triplex Cladding

This all SiC ceramic Triplex fuel clad concept developed by Ceramic Tubular Products (CTP) involves the use of a multilayered (monolith SiC inner layer, SiC CMC middle layer, and outer barrier of CVD SiC) ceramic system to achieve hermeticity needed to retain fission gas and provide a more ductile behavior. The inner layer is a high density monolith to hold fission gases, the intermediate layer is a composite with the required strength, and the outer layer is another dense layer for corrosion protection. As with the SiC/SiC concept, the IAC report noted that maintaining the current pin overall geometry allows for demonstration in existing commercial LWRs, but concerns arise due to the thicker cladding wall, need for higher fuel enrichments, and the need for a robust, hermetic joining. The IAC report noted the need to provide dense impermeable layers while avoiding flaws & cracks and the need for a very tight control of the obviously very complex manufacturing processes. As noted for the SiC/SiC cladding, it is likely that the cladding will be more resistant to fretting, but erosion-corrosion may be an issue [14].

### 3.2.4 Hybrid Cladding: SiC CMC Overbraid onto Zr-based Alloy Tube

The hybrid cladding design is based on a ceramic / metal layered cladding tube utilizing a braided SiC CMC tube as an external wrap onto a commercial Zr-based alloy inner tube (Figure 2). This design utilizes Hi-Nicalon Type S fibers and the PIP process for forming the CMC. In the hybrid design, the hermetic seal for the fuel pin is provided by the inner metal liner and the metal end caps are welded to the inner metal liner using traditional end cap metal to metal joining. The end-cap seal for the fully ceramic system requires sealing of the SiC CMC to itself which has been shown to a major challenge [14]. In addition, the potential effects of Pellet Clad Material Interaction (PCMI) on fuel rod reliability are considered significant with respect to SiC CMC only cladding. GEH proposes that the ceramic / metal hybrid design be considered as an advanced cladding for continued research until further tests and related analyses are available for decisions in the selection of an alternate advanced cladding [12].



**Figure 2. Geometry of SiC CMC Zr-4 hybrid cladding design. A patent for this cladding rodlet concept has been submitted by the Idaho National Laboratory (INL), IDR number BA-477.**

Although excess oxygen from  $\text{UO}_2$  fission reactions is available for oxidation and embrittlement of the inner Zr cladding surface, temperatures at the pellet-cladding interface are sufficiently low under normal LWR operating conditions (400-500 °C) that any cladding oxidation is expected to be minor. However, as with all advanced cladding concepts, reactions with other fission products and reactions under LOCA conditions need further investigation.

### 3.2.5 SiC Development Challenges: Joining

The fully ceramic SiC clad designs require development of a hermetic structure and end-cap seals that can withstand the radiation, temperature and chemical environment inherent to an operating LWR. A reliable, reproducible technique to join and hermetically seal ceramic composites has been identified as a critical technology gap for SiC-based cladding systems. There are a number of conventional and

advanced techniques to join SiC (or SiC/SiC) to itself or other materials. Successfully demonstrated techniques include pre-ceramic polymer joining, glass-ceramics, reaction bonding, active metal / pre-ceramic polymers, and active metal solid state displacement techniques. While the strength of the joints produced by these methods appears to be adequate for LWR applications, there is currently a lack of standards for testing ceramics and a variety of tests have been used to measure the strength of the bonds created using each technique [4].

There is currently limited irradiation data on the joints and materials used to fabricate the joints, and the joint fabrication techniques that have been tested under irradiation have demonstrated poor irradiation stability. Hence, a reliable SiC/SiC joining technique for reactor structural materials has yet to be developed. Given the functional requirement of hermeticity for nuclear fuel cladding, necessary to retain helium and fission products, the SiC/SiC joining technique must be radiation stable for the relevant conditions of applied stress (to be defined), temperature (~400-500°C) and neutron damage (~6 dpa) [4].

Several methods of joining SiC ceramic composites are considered promising for general applications; however, not all are expected to hold promise for in-reactor applications. These methods are summarized in Table 5, along with reported strength properties and anticipated performance under irradiation. Primary considerations for nuclear applications (both fission and fusion) include resistance to neutron irradiation; mechanical properties, such as strength and reliability during mechanical loading; compatibility of the processing condition with the design requirement; chemical compatibility with the operating environment for the intended application; and the ability to satisfy the hermeticity requirement.

**Table 5. Methods for joining SiC-based materials [4].**

<i>Joining Method</i>	<i>Typical Reported Strength</i>	<i>Irradiation Performance</i>
Metal diffusion bonding	~150 MPa shear	Expectedly good
Transient eutectic-phase joining	~250 MPa tensile	Expectedly good
Glass-ceramic joining	~100 MPa shear	Positive result reported (EU program)
Brazing	Various	Generally poor; high activation
SiC reaction bonding	~200 MPa shear	Expectedly unstable
MAX-phase joining	~100 MPa shear	Unknown
Pre-ceramic polymer joining	Tens MPa shear	Expectedly unstable
Transient liquid metal joining	No data	Unknown
Selective area CVD	No data	Expectedly very good

### **3.2.6 SiC Performance: Normal Operation**

#### *Irradiation Stability Criteria*

It is known that at the temperatures of interest for LWR clad there is no effect of flux-rate dependence on damage and the total neutron damage (to stable materials) is the essential factor in evaluating radiation resistance. Without assuming power uprates or extended burn-up fuels, the SiC clad will see approximately 10-15 dpa, which for the nominal 300-400°C operating temperature is well above the saturation condition. It is therefore assumed that a clad structure should be proven stable and resistant to micro cracks up to a minimum 10 dpa to be considered for lead test rod (LTR) deployment. At present, two fiber types (Nicalon Type-S and Tyranno SA) with either graphite or SiC multilayer interphases and CVI SiC matrix have been demonstrated stable beyond these dose and temperature conditions. Stability, as defined in the LWR clad context, is the stability of the overall system, whether a fully ceramic or ceramic / metal hybrid system, to carry out its function within allowables of required strength, swelling, and maximum failure criteria (fission product release) of one rod per million. Presently there is insufficient information pertaining to the stability of any metallic liner / SiC interface or fuel-liner/ fuel-SiC interface [4].

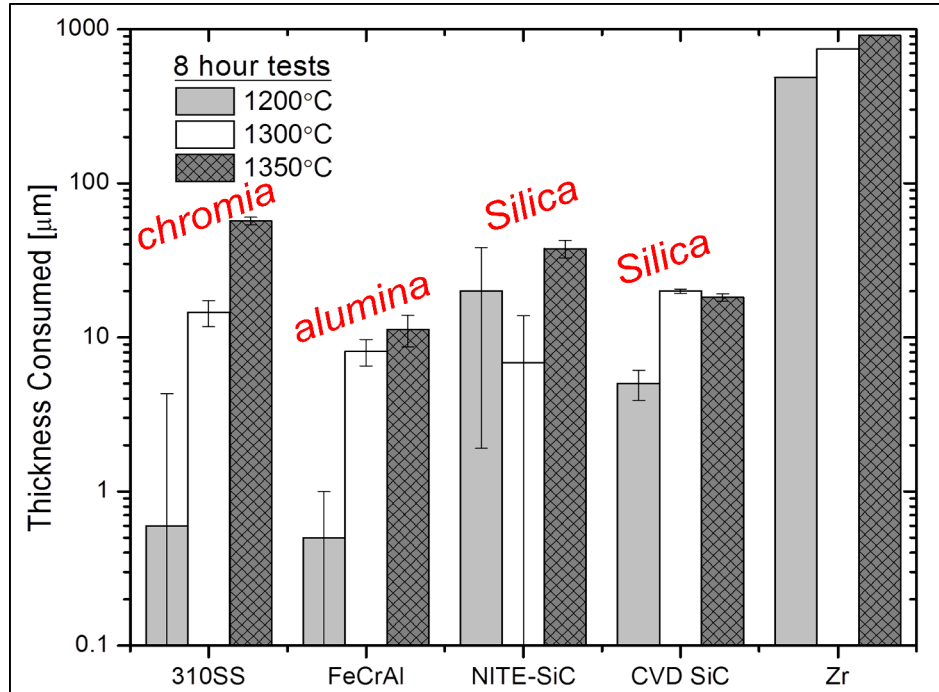
### Corrosion/Erosion

Under normal operating conditions for PWRs (temperature and pressure), pure SiC would be expected to form a semi-protective SiO<sub>2</sub> layer, in effect protecting the surface. Research shows an approximate factor of three difference between a boron sintered SiC ceramic and the better performing CVD SiC for a relatively high velocity 360°C coolant flow. CVD SiC recession has been shown to be on the order of 0.05 mg/cm<sup>2</sup> over a ten day period, or approximately 250 microns over a standard 4.5 year fuel lifetime (without irradiation or appropriate reactor chemistry) [4]. Non-stoichiometric SiC-based materials have demonstrated even more significant mass loss, whether by corrosion or erosion. The mass loss, beyond any concerns regarding irradiation instability underlying the material loss, could raise issues as the very hard SiC particulates (or possibly SiO<sub>2</sub>) are transported through the coolant to heat exchangers and pumps. It is also conceivable that an irradiation-assisted-corrosion process that enhances the surface reaction may occur. In any event, mass loss with regard to secondary system effects and the potential compromise of the mechanical performance of the clad necessitate this as a selection criterion [4].

### **3.2.7 SiC Performance: Off Normal Events**

It is understood that with known oxidation mechanisms and kinetic rates, SiC will react more slowly than zircaloy with steam under LOCA or beyond design basis accident conditions. Near atmospheric pressure the reaction of steam with zircaloy and SiC has been extensively studied and is well understood. While it is well known that metallic materials have a linear pressure dependence of mass loss, this dependence is generally not important over the pressure range associated with reactor transients (for zircaloy) and is hence ignored. However, as SiC has the potential for substantially greater performance than zircaloy it becomes more important to understand the projected pressure/temperature of any beyond design basis accident and the physical mechanism of SiC reactions.

The relative cladding thickness loss of SiC with respect to zircaloy and other candidate cladding materials at both atmospheric and elevated pressures and temperature is now becoming understood as part of various national and international programs. For example, Figure 3 shows the mass loss for the three generic classes of advanced clad: alumina formers, chromia formers, and silica formers, as compared with zircaloy. All the cladding with internally produced oxides outperformed zircaloy, with the CVD SiC showing about two orders of magnitude less thickness consumption at 1200°C. While the outperformance of any SiC-based clad under LOCA and beyond design basis accident conditions is assumed, the relative attractiveness and benefit of the clad (the ultimate economic driver) will depend on the quantitative determination of performance of the clad. For this reason a sufficient understanding of the clad performance under LOCA and/or design base accident conditions is required [4].



**Figure 3. Clad thickness loss for candidate materials in flowing steam at 1 MPa [4].**

SiC cladding appears to have a large potential to mitigate severe accident consequences; however, it is not clear that the material can be successfully manufactured or that it can attain and maintain the current high reliability of existing Zircaloy-clad fuels. In addition, further testing of SiC cladding is needed to examine PCCI, PCMI, degradation of thermal conductivity, ability to form a hermetically sealed rod, and irradiation creep characteristics.

### 3.2.8 Economic Analysis of SiC Cladding

Nuclear electricity production cost consists of two components: operation and management (O&M) costs and fuel costs. A review of historical data from 1995 up to the present points out that the fuel costs in current oxide-fueled nuclear reactors as a share of production cost of nuclear electricity have consistently remained at around 28% [4].

The impact of moving to SiC cladding on the required fuel enrichment will be heavily dependent on the specifics of the chosen fuel design. Causal factors will be any displaced fuel due to a thicker cladding, added absorption associated with the use of a metallic liner/bladder for the case of the hybrid clad design, and potential changes in the extent of neutron moderation due to possible displacement of the water moderator and the presence of SiC. In any event, preliminary estimates (unpublished) indicate a range from essentially no change up to an increased enrichment of ~ 0.5% required [4].

The cost of zirconium alloy cladding in current oxide fuel bundles is roughly \$20k to \$30k per assembly (assuming \$20-\$30 per meter of cladding). Normalized against the mass of LEU in the fuel bundle zirconium alloy cladding cost is 37-55 \$/kgLEU (~\$20-30k/assembly). An estimate to fabricate SiC cladding described in the Advanced LWR Nuclear Fuel Cladding System Development: Technical Program Plan [4] is ~\$900k/assembly, representing a factor of 30 or more increase in the cladding cost. This represents an increase in fuel fabrication cost by a factor of 6.9. Therefore the increase in cost

associated with fabrication of the new SiC-based clad will be the main driver in the overall fuel cost surpassing all other items in magnitude. In the estimate, it was assumed that the current nuclear grade SiC materials have been utilized (such as Nicalon Type-S fibers infiltrated by CVI.) Currently, alternative infiltration methods that may be less costly, such as PIP, are being developed. These methods have undergone limited testing and have not yet resulted in proven irradiation stable products. PIP-fabricated SiC CMCs are therefore considered early in their development phase for irradiation environment application. Additional irradiation testing of SiC CMC materials fabricated via PIP techniques will be required to determine applicability in a reactor environment. Economics may require further investigation of newer, lower cost techniques to ensure that SiC CMC cladding is a viable option. It is also noted that arguments have been made regarding significantly reduced cost of composite constituents (i.e. fibers) as well as the fabrication costs. Current very high production fibers to meet expanding commercial demand for carbon fibers such as polyacrylonitrile (PAN) graphite fibers are approximately 40x less expensive through utilization of much lower cost raw materials [4].

The costs associated with increased cycle lengths are detailed in an Electric Power Research Institute (EPRI) report [3] on PWR cores with SiC cladding. In summary, the use of SiC cladding is expected to reduce the fuel costs by four million dollars per cycle (3-4 percent) with the same number of assemblies loaded as for the Zr-based alloy clad reference core. This savings is primarily due to reduced neutron capture in the carbide compared to Zr-based alloys. As the number of assemblies loaded is reduced to achieve higher discharge burnups, this advantage increases, reaching almost twelve million dollars per cycle (about ten percent) when only 52 assemblies are loaded for a discharge burnup of about 80 MWD/kgU. This burnup level is projected to be quite feasible with SiC. The savings is large enough such it would not be offset by any reasonable fabrication cost increase due to use of SiC. Silicon carbide also allows design of two year cycles with high discharge burnup by loading a half core of fresh fuel. These two-year cycles have similar fuel costs to the reference Zr-based alloy 18-month cycle but offer the advantage of fewer annual refueling outage days on average, providing further savings on the order of five million dollars per year. Two-year cycles using larger numbers of fresh assemblies have smaller discharge burnups, so their fuel costs exceed that of the reference Zr-based core by an amount large enough that it is not compensated by the reduced refueling outage time [3].

### **3.3 $\text{Ti}_3\text{AlC}$ and $\text{Ti}_3\text{SiC}_2$ Triplex Ceramic Cladding (Tubes)**

$\text{Ti}_{n+1}\text{AlC}_n$  belongs to a group of ternary carbides named  $\text{M}_{n+1}\text{AX}_n$  phases (referred to as MAX phase materials). These materials are promising for advanced cladding concepts due to their combination of metallic and ceramic properties; good thermal and electrical conductors, superb machinability, lightweight, thermal shock resistant, damage tolerant, stiff, a relatively low coefficient of thermal expansion, and combine mechanical anisotropy with thermal isotropy [17 and 18]. The WEC report identified both the  $\text{Ti}_3\text{AlC}$  and  $\text{Ti}_3\text{SiC}_2$  triplex ceramics as potential cladding technologies due to machinability and high degree of toughness [1]. Both were rated with the highest thermal conductivity compared to other technologies considered (Zr-based alloys, SiC-based cladding, and 304H stainless steel). Maxthal  $\text{Ti}_3\text{AlC}$  was rated second best (behind SiC) in the WEC study for increased temperature safety margin; however, the WEC noted that these cladding materials had minimal or no development experience [1]. The WEC report also noted that Maxthal ceramic tubes have a severe economic penalty due to their very high thermal neutron capture cross-section, similar to stainless steel.

### **3.4 Coatings – Thermal Spray onto Zr-alloys**

The WEC report identifies thermal spray of  $\text{Ti}_3\text{SiC}_2$  and  $\text{Ti}_3\text{AlC}_2$  or SiC onto Zr alloy as potential cladding technologies that would forestall high temperature corrosion in steam but noted that minimal or no development experience is available.  $\text{Ti}_3\text{AlC}_2$  has good temperature and oxidation resistance which make this material a possible candidate for use as a coating against high temperature oxidation onto Zr-based alloys. The WEC report pointed out that  $\text{Ti}_3\text{AlC}_2$  or SiC sprayed cladding with  $\text{UO}_2$  fuels are

problematic from a private industry perspective due to the long development time required but noted that the spray coatings are attractive due to the low net positive value for nuclear industry developmental worth [1].

The MAX phase coatings have the same favorable properties as the ceramic  $\text{Ti}_3\text{SiC}_2$  and  $\text{Ti}_3\text{AlC}_2$  ceramic tubes for advanced cladding technology. Polycrystalline  $\text{Ti}_3\text{AlC}_2$  is stable in vacuum up to  $1360^\circ\text{C}$ , above which phase decomposition may occur; however, this is much higher than that of Zr-based alloys. The coating thickness is expected to be in the range of 10 to 20-microns. Thermal and cold spray techniques are possible coating techniques. A high film density is required to ensure water and oxygen impermeability. The film provides an additional diffusion barrier for hydrogen and is likely to have positive influence in mitigating hydrogen pickup [14]. Although titanium has a relatively large microscopic neutron absorption cross-section (6.09 barn), the effect of coating on reactivity is expected to be minimal due to the thin layer. It is not clear if a thin film will survive for a sufficiently long duration in a severe accident due to the lack of test data. The IAC report noted that coatings have a large potential to mitigate severe accident consequences by either delaying or reducing the amount of hydrogen generation at elevated temperatures. Since a coating doesn't significantly alter the existing Zircaloy-clad  $\text{UO}_2$  fuel design, a demonstration could be completed in the very near term (5 years or less) provided the necessary materials compatibility information is available. Manufacturing impacts appear to be minimal. No negative impacts on handling or reprocessing were identified by the IAC.

Coatings have a large potential to mitigate severe accident consequences by either delaying or reducing the amount of hydrogen generation at elevated temperatures. Since a coating doesn't significantly alter the existing Zircaloy-clad  $\text{UO}_2$  fuel design, a demonstration could be completed in the very near term (5 years or less) provided the necessary materials compatibility information is available [14]. This type of concept represents the most likely near term accident tolerance enhancement for existing commercial LWRs by the IAC, however, the IAC recommends that a patent study should be completed to determine if the technology already exists in other applications. The IAC also recommends that a search to determine the optimal coating material should also be completed (assuming that the MAX phase material represents one of many candidate materials). [14]

### 3.5 Stainless Steel Tubing

Ferritic-martensitic (F-M) alloys were identified in the GEH report [12] to have the potential to overcome some of the limitations of Zr-based alloys used in fuel rod cladding such as improved corrosion resistance, resistance to hydrogen embrittlement, and high-temperature oxidation resistance. Higher resistance to corrosion and hydrogen-related degradation of properties may provide some limited advantages over Zr-based claddings for design-basis accidents (DBAs) and beyond-design-basis accident (BDBA) conditions where the exothermic reaction of water with Zr-alloys and the resulting hydrogen released from the reaction create concerns with plant safety. These benefits may be offset by potential limitations which include parasitic neutron capture, reduced melting point, adverse dimensional stability, cracking and embrittlement mechanisms. The GEH report [12] recommends technology development to focus on the following:

- Performance potential assessment based on thermal-mechanical evaluations using presumed properties of F-M alloy cladding, incorporating benefits of higher strength and oxidation resistance and disadvantages related to a lower melting temperature than for Zr-based alloys.
- Assessment of the economic benefits or disadvantages associated with F-M alloy cladding, which can be determined relative to Zr-based cladding. This should include fuel cycle economic assessment as well as any financial benefits for reactor operation associated with safety benefits of this cladding material.
- If the economic assessment shows that work development is warranted, then corrosion performance of F-M alloys in LWR conditions should be assessed.

- If the corrosion assessment shows that beneficial corrosion performance is attainable, then work should continue with alloy selection based on optimization of properties.

Since ferritic-martensitic steel materials have mainly been developed in the sodium fast reactor application, a successful cladding material will need sufficient evaluation and testing to show that the previously demonstrated irradiation assisted stress corrosion cracking (IASCC) issues associated with these materials in the sodium fast reactors are not present in LWR environments. Additionally, the thermal and irradiation creep properties are key characteristics to enable sufficiently high power production, since this is one of the advantageous parameters thought to be provided by such materials. In the process of assessing the material application to a particular type of design, a trade-off study should be performed to assess whether the strength and corrosion tolerance can be used to augment power and to offset the worsening of the neutron economy associated with Fe and alloying elements [11].

Additional engineered stainless steel (SS) alloy concepts are being developed as fuel cladding technologies. These SS alloys include ferritic Oxide Dispersion Strengthened (ODS) stainless steels and austenitic stainless steels [19]. Some of the promising properties of these SS alloys include [19]:

- Oxidation resistance to 1300 °C in steam
- Water corrosion resistance
- High temperature strength
- Ductility-toughness
- Improved thermal neutron cross-section
- Fabricability (austenitic and ferritic/martensitic alloys are easiest to work with - techniques are being developed and tested for ODS alloys)
- Joining using conventional methods (welding of thin walled tubing needs to be developed)
- Radiation tolerance (needs to be investigated under LWR irradiation conditions)

### 3.5.1 Advanced Zr-alloys

The GEH assessment report [11] noted that fuel fabricators and plant operators in other countries, such as Japan or Korea, benefit from national programs developing advanced Zr alloys for LWR application. Advanced Zr-alloy cladding materials would be most easily assimilated into the current licensing methods and regulatory framework due to their similarity to and improvement upon existing Zr-alloys. The primary benefits of such an alloy would come through improved corrosion resistance and reduced hydrogen-related impact on properties (i.e., reduced hydrogen pickup and/or increased hydrogen solubility to reduce formation of zirconium hydrides).

GEH has proposed that the advanced Zr-alloys (or set of alloys) meet the following objectives relative to Zr-2 and other current BWR alloys:

- Reduced corrosion rate
- Reduced hydrogen pickup
- Increased hydrogen solubility, or improved hydrogen-affected properties
- Reduced high-temperature oxidation rate
- Irradiation growth behavior similar to Zr-2, Zr-4 and/or Zr-Nb alloys
- High-temperature strength and creep strength similar to Zr-2
- Fabrication characteristics similar to current Zr alloys, or economically viable

The primary safety benefits would come from improvements to corrosion and hydriding that would increase resistance to fuel failure during DBAs. Reduction of elevated temperature corrosion rate would increase resistance to cladding degradation and perforation under LOCA conditions. Reduction of



H<sub>2</sub> pickup and/or improvement of hydrogen-affected properties will improve post-quench ductility and increase margins to safety. Margins to RIA-induced breach would be increased by an alloy with resistance to hydrogen-related degradation of properties. A cladding alloy meeting the objectives above is not likely to allow significant increases in LHGR or in bundle power unless the current limitation preventing dry-out is removed or relaxed. However, improvements in corrosion resistance and hydrogen resistance would enhance fuel burnup capability. Sourcing and manufacturing of an advanced Zr-alloy would likely have similar cost to current Zr-alloys, except perhaps the lower volume of an advanced Zr-alloy preventing benefits that come with larger orders. Improvement to corrosion resistance and to hydrogen-related properties (like ductility) would improve fuel rod failure resistance under storage conditions. Sustained operation at the higher burnup attainable with the envisioned cladding alloy would eventually reduce somewhat the number of fuel bundles discharged into storage or sent to a fuel recycle process. The fuel recycle process would be unchanged by use of advanced Zr-alloy cladding. The potential benefit of an advanced Zirconium-based alloy is derived from the ability to have a hydrogen pickup fraction below the solubility limit in order to prevent hydride formation. While there are possible candidate formulations for such a material, it is not known that this objective is attainable. That essential characteristic needs to be balanced with a low oxidation rate at elevated temperatures: The key area of concern is whether these characteristics can be made sufficiently low to make a significant change compared to Zircaloy characteristics. As with all cladding materials, the creep and growth characteristics of the more corrosion resistant alloy will need to be balanced with the other key characteristics. [11]

## 4. CONCLUSIONS

Preliminary fuel cladding technology performance criteria and cladding technologies have been identified through an in-depth nuclear industry literature search. A performance criteria table was constructed categorizing these criteria into three groups: critical performance criteria, desired performance criteria, and licensing criteria. Weights will eventually be assigned to each criterion based on input from stakeholders, nuclear industry experts, and the nuclear material science community. Leading candidate technologies will be evaluated against one another based on these weighted parameters and a selection will eventually be made based on this evaluation for further development and testing. A comparison between leading candidate cladding technologies is presented emphasizing the potential benefits and drawbacks for each of the technologies. A discussion on the nuclear design trade-offs, accident performance, storage, economics, safety, and licensing is offered for each candidate technology as a basis for the performance criteria and technology selections.

The leading candidate technologies identified in this Trade-off Study include coated zircalloys, SiC/SiC, SiC CMC Zr-based alloy hybrids, advanced zircalloys, and engineered stainless steel alloys. A technical and economic screening of multiple fuels, claddings, and geometries performed by WEC [1] found that SiC cladding offers the best economic performance and was the highest rated cladding for increased temperature safety margin compared to the other cladding options considered in the study (Maxthal Ti<sub>3</sub>SiC<sub>2</sub>, Maxthal Ti<sub>3</sub>AlC<sub>2</sub>, thermal spray coated Zr, and 304H stainless steel); however, the WEC report did not distinguish between the monolithic and fiber reinforced ceramic matrix composite (CMC) forms of SiC for use as cladding. However, there are concerns with SiC based cladding that require further investigation. In particular, the fuel-cladding gap in the SiC closes much more slowly than in Zr-based alloys due primarily to the lack of creep in SiC [3]. Changing the gap size, increase in fission gas pressure, and the use of annular fuel with a 10% void space is a reasonable solution to this concern by significantly reducing the maximum fuel temperature. Unfortunately, this would also reduce the mass of uranium loaded by 10% requiring an increase in fuel enrichment [3]. Also, a reliable, reproducible technique to join and hermetically seal ceramic composites has been identified as a critical technology gap for SiC-based cladding systems. Additionally, it is not clear how the material will perform in normal operating conditions if subjected to assembly distortion or fuel rod bow behaviors. Additional research is also needed to evaluate potential fission product reactions and PCCI and PCMI.

GEH proposes that the U.S. DOE consider a long-term cladding development program that can include investigation and development of SiC, stainless steel or ferritic-martensitic alloys and advanced zirconium alloys [12]. The primary objectives would be cladding materials with improved performance properties and significantly improved behavior under severe accident conditions, but additional objectives can include advanced zirconium alloys that offer greatly improved behavior under normal operating conditions and DBAs while maintaining high temperature oxidation behavior common to Zr-alloys under severe accident conditions.

The IAC report noted that coated Zr-based alloys represents the most likely near term accident tolerance enhancement for existing commercial LWRs, however, the IAC recommends that a patent study should be completed to determine if the technology already exists in other applications. The IAC also recommends that a search to determine the optimal coating material should also be completed (assuming that the MAX phase material represents one of many candidate materials) [14].

In summary, although all the technologies identified in this study have potential benefits and drawbacks (Table 40, the information provided in this Trade-off Study can be used in conjunction with continued input from industry stakeholders, national laboratories, and the U.S. Department of Energy-Nuclear Energy (DOE-NE) to identify technologies warranting further development and testing through the LWRS program. It is noted that additional technologies not included in this study may also warrant further study, however, this study was based on the most mature technology development and testing available through nuclear industry reporting.

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